

REMARKS

The specification is amended in order to claim benefit of prior-filed applications.

Claims 1-8 and 15-20 are cancelled without prejudice.

Claims 9 and 11-14 are amended.

Claim 10 is as originally presented.

Claims 21-27 are added.

The amendment to the claims serves to remove multiple dependencies and to improve readability.

Support for the amendment to claim 9 is found in original claims 1 and 9.

Support for new claims 21-26 is found in original claims 15-20 and throughout the specification as originally filed.

Support for new claim 27 is found in original claim 11.

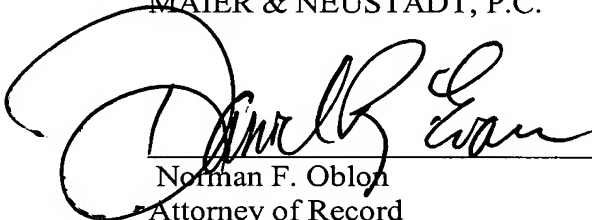
Therefore, no new matter will be entered upon entry of the amendment.

Upon entry of the amendment claims 9-14 and 21-27 will be active.

An action on the merits and allowance of the claims is requested.

Respectfully submitted,

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Process for the preparation of surfactant alcohols and surfactant alcohol ethers, the prepared products and their use

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The present invention relates to a process for the preparation of surfactant alcohols and surfactant alcohol ethers which, inter alia, are highly suitable as surfactants or for the preparation of surfactants. In the process, starting from C₄-olefin streams, olefins or olefin mixtures are prepared by a metathesis reaction which are dimerized to give an olefin mixture having from 10 to 16 carbon atoms, which comprises less than 10% by weight of compounds which have a vinylidene group, then the olefins are derivatized to give surfactant alcohols and said alcohols are optionally alkoxyated.

15 The invention further relates to the use of the surfactant alcohols and surfactant alcohol ethers for the preparation of surfactants by glycosylation or polyglycosylation, sulfation or phosphorylation.

20 Fatty alcohols having chain lengths from C₈ to C₁₈ are used for the preparation of nonionic surfactants. They are reacted with alkylene oxides to give the corresponding fatty alcohol ethoxylates. (Chapter 2.3 in: Kosswig/Stache, "Die Tenside" [Surfactants], Carl Hanser Verlag, Munich Vienna (1993)). The chain length of the fatty alcohol influences the various surfactant properties, such as, for example, wetting ability, foam formation, ability to dissolve grease, cleaning power.

25 Fatty alcohols having chain lengths from C₈ to C₁₈ can also be used for preparing anionic surfactants, such as alkyl phosphates and alkyl ether phosphates. Instead of phosphates, it is also possible to prepare the corresponding sulfates. (Chapter 2.2. in: Kosswig/Stache "Die Tenside" [Surfactants], Carl Hanser Verlag, Munich Vienna (1993))

30 ~~Such fatty alcohols are obtainable from native sources, e.g. from fats and oils, or else in a synthetic manner by constructing building blocks having a lower number of carbon atoms. One variant here is the dimerization of an olefin to give a product having twice the number of carbon atoms and its functionalization to give an alcohol.~~

35 For the dimerization of olefins, a number of processes are known. For example, the ~~reaction can be carried out over a heterogeneous cobalt oxide/carbon catalyst~~

amended page 2

as enclosed to IPER

DE-A-196 04 466 is concerned with aqueous compositions containing an alkylglycoside and a polyethyleneglycol derivative of formula I given in this document.

- 5 The alkyl group R^2 (Page 2, line 55) has 8 to 18, preferably 10 to 16 carbon atoms; no direct information is given in this document about the degree of branching. One can, however, conclude that the alkyl group must be predominantly linear, because it is said that it has been obtained by hydrogenation of native fatty acids.
- 10 Such fatty alcohols are obtainable from native sources, e.g. from fats and oils, or else in a synthetic manner by constructing building blocks having a lower number of carbon atoms. One variant here is the dimerization of an olefin to give a product having twice the number of carbon atoms and its functionalization to give an alcohol.
- 15 For the dimerization of olefins, a number of processes are known. For example, the reaction can be carried out over a heterogeneous cobalt oxide/carbon catalyst (DE-A-1 468 334), in the presence of acids such as sulfuric or phosphoric acid (FR 964 922), with an alkyl aluminum catalyst (WO 97/16398), or with a dissolved nickel complex catalyst (US-A-4 069 273). According to the details in US-A-4 069 273, the use of these nickel complex catalysts (the complexing agent used is 1,5-cyclooctadiene or
- 20 1,1,1,5,5,5-hexafluoropentane-2,4-dione) gives highly linear olefins with a high proportion of dimerization products.

- DE-A-43 39 713 (D1) is concerned with a process of oligomerization of olefins using catalysts, which have been tailored so that there are obtained extraordinary high
- 25 proportions of linear reaction products, which are particularly desired with this process.

Working Examples 3 to 5 of this document shows oligomerization of butan/butene-mixtures, whereby reaction products are obtained containing 62 to 78 % by weight of Octen. This known process comprises no metathesis and the reaction products disclosed therein do not consist of components having 10 to 16 carbon atoms.

amended page 2a

as enclosed to IPER

US-A-3,448,163 (D3) is concerned with a process for diproportionation of olefins and catalysts, which are particularly useful for this process. In the Working Example there is shown that butene-1 is transformed into a mixture of olefins having 2 to 7 carbon atoms, particularly ethylene and hexene-3. this known proces comprises no dimerisation step and the reaction product disclosed therein does not consist of components having 10 to 16 carbon atoms.

Functionalization of the olefins to give alcohols with construction of the carbon skeleton about a carbon atom expediently takes place via the hydroformylation reaction, which gives a mixture of aldehydes and alcohols, which can then be hydrogenated to give alcohols. Approximately 7 million metric tons of products per annum are produced worldwide using the hydroformylation of olefins. An overview of catalysts and reaction conditions for the hydroformylation process are given, for example, by Beller et al. In Journal of Molecular Catalysis, A104 (1995), 17-85 and also in Ullmann's Encyclopedia of Industrial Chemistry, vol. A5 (1986), page 217 et seq., page 333, and the relevant literature references.

GB-A-1 471 481 (D2) is concerned with a process for hydroformylation olefins using a catalyst containing cobalt. The olefins introduced in this process are linear and, hence, oxoalcohols an oxoaldehydes are obtained having a low degree of branching.

From WO 98/23566 it is known that sulfates, alkoxylates, alkoxysulfates and carboxylates of a mixture of branched alkanols (oxo alcohols) exhibit good surface activity in cold water and have good biodegradability. The alkanols in the mixture used have a chain length of greater than 8 carbon atoms, having on average from 0.7 to 3 branches. The alkanol mixture can, for example, be prepared by hydroformylation, from mixtures of branched lefins which for their part can be obtained either

as originally filed - 2 -

~~(DE-A 1 468 324), in the presence of acids such as sulfuric or phosphoric acid (EP 964 922), with an alkyl aluminum catalyst (WO 97/16398), or with a dissolved nickel complex catalyst (US-A-4 069 273). According to the details in US-A-4 069 273, the use of these nickel complex catalysts (the complexing agent used is~~
 5 ~~1,5-cyclooctadiene or 1,1,1,5,5,5-hexafluoropentane-2,4-dione) gives highly linear olefins with a high proportion of dimerization products.~~

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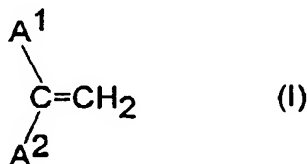
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 25 ~~obtained either by skeletal isomerization or by dimerization of internal, linear~~ olefins.

A given advantage of the process is that a C₃- or C₄-olefin stream is not used for the preparation of the dimerization feed. It follows from this that, according to the current prior art, the olefins subjected to dimerization therein must have been prepared from ethylene (e.g. SHOP process). Since ethylene is a relatively
 30 expensive starting material for surfactant manufacture, ethylene-based processes have a cost disadvantage compared with processes which start from C₃- and/or C₄-olefin streams.

Another disadvantage of this known process is the use of mixtures of internal olefins, which are only obtainable by isomerization of alpha-olefins, which is
 35 required for the preparation of branched surfactant oxo alcohols. Such processes always lead to isomer mixtures which, because of the varying physical and chemical data of the components, are more difficult to handle in terms of process

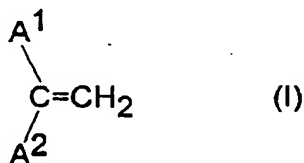
CLAIMS as enclosed to IPER

- 5 1. A process for the preparation of surfactant alcohols and surfactant alcohol ethers by derivatization of olefins having from about 10 to 20 carbon atoms or of mixtures of such olefins and optionally subsequent alkoxylation, which comprises
- 10 a) subjecting a C₄-olefin mixture to metathesis,
b) separating off olefins having from 5 to 8 carbon atoms from the metathesis mixture,
c) subjecting the separated-off olefins individually or as a mixture to dimerization to give olefin mixtures having from 10 to 16 carbon atoms,
d) subjecting the resulting olefin mixture, optionally after fractionation, to derivatization to give a mixture of surfactant alcohols, and
e) optionally alkoxyating the surfactant alcohols.
- 15 2. A process as claimed in claim 1, wherein the process step a), the metathesis, is carried out in the presence of catalysts containing molybdenum, tungsten or rhenium.
- 20 3. A process as claimed in one of claims 1 and 2, which comprises, in process step b), separating off the olefins having 5 and 6 carbon atoms.
4. A process as claimed in one of claims 1 to 3, wherein process step c), the dimerization, is carried out with heterogeneous catalysis.
- 25 5. A process as claimed in one of claims 1 to 4, wherein a dimerization catalyst is used which contains at least one element from subgroup VIII of the Periodic Table of the Elements,
- 30 and the catalyst composition and the reaction conditions are chosen such that a dimer mixture is obtained which comprises less than 10% by weight of compounds which have a structural element of the formula I (vinylidene group)



in which A¹ and A² are aliphatic hydrocarbon radicals.

6. A process as claimed in one of claims 1 to 5, wherein, in process step c) olefins having 5 and 6 carbon atoms are dimerized individually or in a mixture.
- 5 7. A process as claimed in one of claims 1 to 6, wherein, in process step c), 3-hexene is dimerized.
8. A process as claimed in one of claims 1 to 7, wherein the derivatization (process step d)) is carried out by hydroformylation.
- 10 9. A novel olefin mixture preparable by process steps a), b) and c) of the process of claim 1, wherein
- a) the components have from 10 to 16 carbon atoms
- b) the proportion of unbranched olefins is less than 25% by weight
- 15 c) the proportion of components having a structural element of the formula I (vinylidene group)



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wherein A1 and A2 represent aliphatic hydrocarbon groups, is below 10 % by weight

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10. An olefin mixture as claimed in claim 9, which has a proportion of unbranched olefins of less than 20% by weight.
- 30 11. An olefin mixture as claimed in one of claims 9 and 10, wherein at least 80% of the components of the dimerization mixture have, in the range from 1/4 to 3/4, preferably from 1/3 to 2/3, of the chain length of their main chain, one branch, or two branches to adjacent carbon atoms.
- 35 12. An olefin mixture as claimed in one of claims 9 to 11, wherein, at the branching sites of the main chain, predominantly groups having (γ-4) and (γ-5) carbon atoms

are bonded, where y is the number of carbon atoms in the monomer used for the dimerization.

13. An olefin mixture as claimed in one of claims 9 to 12, wherein the ratio of aliphatic to olefinic hydrogen atoms is in the range
5 $H_{\text{aliph.}} : H_{\text{olefin.}} = (2 \cdot n - 0.5) : 0.5$ to $(2 \cdot n - 1.9) : 1.9$, where n is the number of carbon atoms in the olefin obtained in the dimerization.
14. An olefin mixture as claimed in one of claims 9 to 13, wherein the ratio of aliphatic to olefinic hydrogen atoms is in the range
10 $H_{\text{aliph.}} : H_{\text{olefin.}} = (2 \cdot n - 1.0) : 1$ to $(2 \cdot n - 1.6) : 1.6$.
15. Surfactant alcohols preparable by the process steps a), b), c), d) and optionally e) of the process of claim 1,
15 a) having 11 to 17 carbon atoms and
b) comprising a proportion of unbranched alcohols of below 25 % by weight, and its alkoxylation products.
16. The use of the surfactant alcohol alkoxylation products of claim 15 as nonionic
20 surfactants.
17. The use of the surfactant alcohol of claim 15 for the preparation of surfactants.
18. The use of the surfactant alcohol of claim 15 for the preparation of alkanol
25 glycoside and polyglycoside mixtures by single or multiple reaction (glycosylation, polyglycosylation) with mono-, di- or polysaccharides with the exclusion of water and with acid catalysis or with O-acetylsaccharide halides.
19. The use of the surfactant alcohol and its alkoxylation products of claim 15 for the
30 preparation of surface-active sulfates by esterification thereof with sulfuric acid or sulfuric acid derivatives to give acidic alkyl sulfates or alkyl ether sulfates.
20. The use of the surfactant alcohol and its alkoxylation products of claim 15 for the
35 preparation of surface active phosphates by esterification thereof with phosphoric acid or its derivatives to give acidic alkyl phosphates or alkyl ether phosphates.